Tracing the Sources and History of Subsurface Contamination at the Hanford Site in Washington Using High-Precision Uranium Isotopic Measurements

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Groundwater contamination at the Hanford Site, Washington, resulted from decades of nuclear fuel production and processing. Understanding the fate and transport of contaminants has been complicated by the presence of multiple potential sources within relatively small areas. The contrasts in isotopic composition between natural and anthropogenic uranium promotes the measurement of uranium isotopic composition as a fingerprint and tracer of uranium contamination in the environment. In this study we focused on a uranium groundwater plume and two vadose zone U plumes associated with the B-BX-BY waste management area, used since the late 1940's for the storage of high-level radioactive waste. Groundwater U contamination was first detected in monitoring wells in the early 1990's. The groundwater plume ($>$30 ppb U) is currently approximately 250 m wide, at least 900 m long with U concentrations up to 525 ppb. The exact origin and history of this contamination is not well understood, since a number of tanks and incidents are potential sources. Using multiple collector ICPMS (Micromass IsoProbe) high precision uranium isotopic analyses were conducted of samples of vadose zone contamination and of groundwater. The measured isotopic compositions (^{234}U/^{238}U, ^{235}U/^{238}U and ^{236}U/^{238}U) are used to distinguish contaminant sources, and are compared to a history of processed U fuel rod compositions. Based on the isotopic data, the groundwater plume appears to be related to a tank overflow event in 1951 that spilled high-level waste into the vadose zone. A second zone of identified vadose zone contamination does not seem to be an important contributor to the groundwater U plume. The variation in U isotopic composition of the groundwater samples is a result of mixing of contaminant U from the 1951 event and natural background U. The identified vadose zone contamination source is horizontally displaced from the initial locus of groundwater contamination, indicating that lateral migration within the vadose was at least 8 times greater than vertical migration. The time evolution of the groundwater plume suggests an average U migration rate of $\sim$0.7 m/day showing slight retardation relative to groundwater flow of $\sim$1 m/day.